

Theoretical Analysis of the Propagation of a Reaction Front in a Packed Bed

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The propagation of a reaction front through a packed bed is analyzed theoretically. The chemical reaction rate is represented by Arrhenius temperature kinetics with external transfer limitation and general power law dependency on both gaseous and solid reactant concentrations. Analogous to so-called "Activation Energy Asymptotics" developed for premixed laminar flames, the largeness of the activation energy of the chemical reaction is exploited to derive asymptotic solutions from the three governing differential equations pertaining to transport of heat, of solid reactant, and of gaseous reactant, making use of the method of matched asymptotic expansions. Two regions are distinguished, i.e., an outer region or preheat zone and an inner region or reaction zone. In the preheat zone, the reaction terms can be neglected as compared to the convective and diffusive terms. In the reaction zone, the diffusion of heat is dominating over the convective heat transport mechanism and balances the heat of reaction. In accordance with the magnitudes taken for the Lewis numbers, for solid and gaseous reactants convective transports are dominating and submitting diffusive transports in the reaction zone, respectively. Solutions in closed form are presented for governing variables including reaction front velocity whereby previously published results appear as special cases. The solutions provide direct insight into underlying physical processes and enable the effects of important parameters to be quantified analytically.

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NOMENCLATURE

B_0	integration constant
C_0	integration constant
C_p	apparent specific heat [J/(kg K)]
c_p	specific heat [J/(kg K)]
$D_{a,eff}$	effective axial dispersion coefficient [m ² /s]
E_a	activation energy [J/mol]
E_s	dispersion coefficient for solid phase reactant [m ² /s]
ΔH_r	reaction enthalpy [J/kg]
K_g	ratio of generalized transport coefficient to reaction rate at temperature T^b
K_r	reaction rate at temperature T^b [(m ³ /kg) ^{$m+n-1$} s ⁻¹]
k_o	generalized pre-exponential factor [(m ³ /kg) ^{$m+n-1$} s ⁻¹]
k_g	generalized transport limitation coefficient [(m ³ /kg) ^{$m+n-1$} s ⁻¹]
k_r	reaction rate at temperature T [(m ³ /kg) ^{$m+n-1$} s ⁻¹]
Le	effective Lewis number
m	reaction order solid key reactant
n	reaction order gaseous key reactant

q	diffusive heat flux [W/m ²]
R	universal gas constant [J/(mol K)], total reaction rate [kg/(m ³ s)]
T	temperature [K]
u_f	reaction front velocity [m/s]
U_g	superficial gas velocity [m/s]
x	space coordinate in moving coordinate system [m]
X	scaled dimensionless space coordinate
y	mass fraction

Greek Symbols

α	scaled dimensionless "burnt" mass fraction of gaseous key reactant
$\Gamma(v)$	complete gamma-function
$\Gamma(v, w)$	incomplete gamma-function
γ	Zeldovich number
δ	arbitrary dimensionless number
ϵ_g	packed bed porosity
ζ	scaled mass fraction of gaseous key reactant
η	inner coordinate, effectiveness factor
θ	dimensionless temperature
κ_r	scaling constant for asymptotic analysis [W/(m ² K)]

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$\Lambda_{a,eff}$	effective packed bed axial heat conduction coefficient [W/(m K)]
μ	relative temperature rise
v_i	stoichiometric factor
Φ	convective mass flow [kg/(m ² s)]
ϕ	dimensionless convective energy flux
ξ	notation for Riemann's zeta-function
ρ	density [kg/m ³]
σ	scaled mass fraction of solid key reactant
ψ	dimensionless diffusive heat flux

Frequently Used Indices

<i>b</i>	“burnt” situation, situation after reaction is completed
<i>c</i>	convection dominated
<i>d</i>	diffusion dominated
<i>g</i>	related to gaseous phase
<i>i</i>	<i>i</i> th reactant, here: key reactant in gaseous phase
<i>in</i>	inner zone or reaction zone
<i>k</i>	<i>k</i> th reactant, here: key reactant in solid phase
<i>ou</i>	outer zone or preheat zone
<i>s</i>	related to solid phase
<i>u</i>	“unburnt” situation, initial situation before reaction

INTRODUCTION

Many chemical conversion processes are carried out in (catalytic or noncatalytic) packed bed reactors or in homogeneous plug flow. If the cold reactants are separated from the high-temperature reaction products by a relatively thin area in which the reactions take place, this area is called the reaction front. The reaction front propagates with a certain relative velocity through the medium. For purpose of illustration, the premixed combustion in laminar flames, the production of ceramic materials by combustion synthesis processes, the conversion of chemical reactants in catalytic packed beds, and the fixed bed combustion or gasification of solid fuels such as municipal waste and coal can be mentioned [1].

At the initial temperature the reactant is in a basically nonequilibrium thermodynamic state, but chemical reactions are extremely slow. Heat

generated in the intense chemical conversion zone is conducted to the upstream cold layers, heating them and increasing the reaction rate. As a result, propagation of the exothermic reaction front occurs. In addition to heat conduction, diffusion of components over distances comparable to the reaction front thickness possibly occurs. The reaction front propagates into the reactant mixture with the reaction front propagation speed or simply the front speed u_f . In case of a premixed flame, the front speed is called the laminar flame speed and the problem is known as the freely propagating flame problem. When the temperature dependency of the chemical reaction is large enough, two zones can be distinguished within the reaction front, the preheat zone and the reaction zone. In the preheat zone, the effect of the reaction can be disregarded. In the reaction zone, the chemical reaction is intense and can not be neglected. The temperature rise from the initial temperature to temperatures somewhat lower than the maximum reaction temperature occurs at the expense of heat conducted from the reaction zone to (and within) the preheat zone.

In this paper, a theoretical analysis of the propagation of a reaction front in a packed bed is presented. The analysis concentrates on a situation in which the reaction front moves in opposite direction with respect to the gas flow through the packed bed. For the solution of the governing differential equations, use has been made of the “method of matched asymptotic expansions” [2]. Although this method is capable of producing useful results of a high order of accuracy, it is not either its only or its most notable feature. Just the acquisition of a complete first approximation in analytical form produces such a gain in understanding of the physics of a situation that this alone makes the method extremely valuable [3].

Most studies on reaction front propagation problems are involved with the freely propagating laminar flame or burner stabilized flame problem, in which diffusion of the gaseous reactants is of the same order as the conduction of heat (i.e., the Lewis number is equal to unity or of order unity) [4–6]. The limiting problem in which diffusion can be disregarded or is absent, i.e., only convective transport of the reactants is important, is considered numeri-

cally by Von Karman [4] and analytically by Merzhanov and Khaikin [5] and Gatica et al. [7]. In contrast with a gas flame where the main concentration change occurs in the preheat zone due to diffusion of reactants and conduction of heat, the main change in concentration in a medium with no or very low diffusion occurs in the reaction zone. This is due to a negligible role of diffusive transfer on distances of the order of the reaction zone width. More recent applications of the method in the field of packed bed combustion are given by Dosanjh et al. [8], Fateni and Kaviany [9], and Escobedo and Viljoen [10]. A packed bed can be considered as a combination of the diffusion governed and the convection governed case.

In this paper an analysis based on large activation energy asymptotics is performed taking account of two reactants simultaneously. Whereas for the gaseous reactant diffusion is dominant in the reaction zone, for the solid reactant convective transport dominates. Solutions in closed form are given for governing variables whereby previously published results appear as special cases. Furthermore, an approach is proposed for cases that are not governed by either limiting situation.

The results can be applied to any packed bed conversion process where a reaction front is formed that moves in opposite direction to the flow of the gaseous reactant, or to the simplified case of pure homogeneous (either gaseous or in solid state) conversion processes. Although several assumptions have been made with regard to the functional shape of the reaction rate equation, the analysis can equally well be applied to any reaction where the reaction rate close to the maximum temperature is exponentially large as compared to the reaction rate at lower temperatures. Furthermore, the analysis assumes that the diffusive processes can be described by dispersion coefficients that can take the shape of a constant for homogeneous processes or are a function of the bed properties and flow parameters for a heterogeneous packed bed [1, 11, 12]. The impact of the various assumptions will be discussed where relevant. An assessment of the physical value of the parameters for various applications, as well as a comparison to experimental results is presented elsewhere [1].

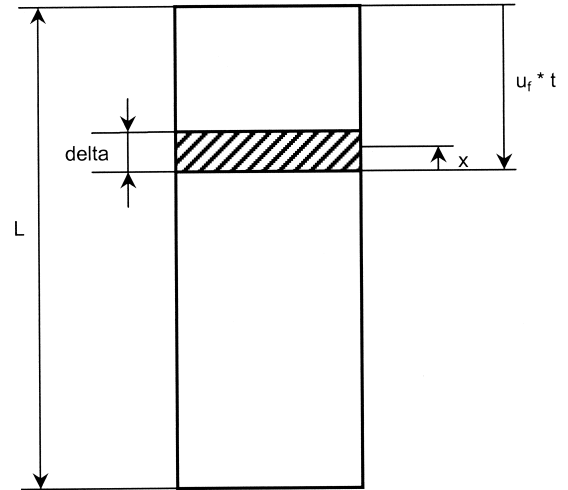


Fig. 1. Schematic representation of the propagation of a reaction front in a packed bed. Illustration of coordinate transformation.

GOVERNING EQUATIONS

The situation of a reaction front, propagating in a packed bed of reacting particles, is schematically illustrated in Fig. 1. The situation is considered here, where the reaction front moves in opposite direction with respect to the gaseous flow entering from below.

Adopting a pseudohomogeneous modeling approach and a one-step global heterogeneous reaction with a key reactant in the solid phase and a key reactant in the gaseous phase, the governing equations can be written as [1]:

$$-u_f \rho_s \frac{\partial y_k}{\partial x} + \frac{\partial}{\partial x} \left(\rho_s E_s \frac{\partial y_k}{\partial x} \right) - R = 0, \quad (1)$$

$$-U_g \rho_g \frac{\partial y_i}{\partial x} + \frac{\partial}{\partial x} \left(\rho_g D_{a,eff} \frac{\partial y_i}{\partial x} \right) - v_i R = 0, \quad (2)$$

$$-(U_g \rho_g c_{p_g} + u_f \rho_s c_{p_s}) \frac{\partial T}{\partial x} + \frac{\partial}{\partial x} \left(\Lambda_{a,eff} \frac{\partial T}{\partial x} \right) + R(-\Delta H_r) = 0 \quad (3)$$

where the coordinate system is attached to the reaction front, moving at a constant front speed u_f . The equations can alternatively be obtained from the full instationary equations, by transforming to a coordinate system attached to the reaction front and neglecting transient terms,

which are of order $O(\delta/L)$ compared to the time needed for complete conversion of a layer with thickness L , with δ the reaction front thickness and generally $\delta \ll L$.

The effective dispersion coefficients for mass and energy can be described as a functional dependency of various physical parameters [1, 11, 12]. The effective mass diffusion coefficients E_s and $D_{a,eff}$ are either constant or a function of bed properties and flow parameters only. The effective thermal conductivity $\Lambda_{a,eff}$ also depends on the bed temperature by means of the radiative contribution, especially at high conversion temperatures. It appears, however, that the contribution due to fluid flow largely exceeds the radiative contribution at higher fluid velocities. Therefore, the packed bed effective heat conductivity coefficient is assumed constant and is evaluated at the mean conversion temperature. This approach can be shown correct in first order as long as the flow velocities are not $O(10^{-2})$ or below, by means of an asymptotic analysis [1].

The momentum equations are bypassed by assuming isobaric and inviscid flow. Furthermore, in Eqs. 1 to 3, y_i and y_k are used to denote the mass fraction of the gaseous (g) and solid-phase (s) reactants respectively. The solid density in the packed bed ρ'_s is the product of the local density ρ_s^o within the solid phase and a factor $(1 - \epsilon_g)$ representing the fraction of volume filled with solid material. The fraction y_k incorporates the combined effect of changing solid density in the packed bed and changing fraction y'_k of the key reactant in the solid phase:

$$y_k = \frac{\rho'_s}{\rho_s} * y'_k, \quad (4)$$

where ρ_s is a constant, representing the packed bed solid density before reaction.

A generalized description of the effective reaction rate is adopted, which is n th order in the gaseous key reactant and m th order in the solid key reactant:

$$R = \frac{1}{\frac{1}{k_g} + \frac{1}{k_r}} (\rho_s y_k)^m (\rho_g y_i)^n, \quad (5)$$

$$k_r = k_o * e^{-E_a/(R*T)}. \quad (6)$$

Equation 5 is discussed in detail elsewhere [1]. The constant k_g constitutes a generalized limitation coefficient, to account for effects of mass or heat transfer to the external reaction surface. The pre-exponential factor k_o and activation energy E_a may be lumped kinetic parameters, i.e., for fully internally limited reactions in combusting coal particles the lumped activation energy appears as half of the true Arrhenius value [13, 14], but could vary anywhere between 0.5 and 1 times the true value depending on the extent of diffusional control [14]. The description of the reaction rate is exact in case of true Arrhenius kinetics, i.e., $k_g/k_r \gg 1$, and in case of a first-order partially external mass transfer limited reaction. For all other cases an explicit reaction can not be derived, e.g., for a n th order partially external mass transfer limited reaction an implicit rate equation is obtained [15]. In such a situation, the reaction rate can only be obtained by iterative methods. Thus, in general, consecutive rate processes of general order can not easily be described by an overall explicit expression. Similar conclusions can be drawn for nonisothermal situations [15–17]. In all these cases, the use of a generalized rate equation (5) may provide a means to describe the reaction rate by a matching explicit equation. Although the proposed rate equation is defined as general as possible, situations may arise where the resulting functional form is not satisfying. In such a case, the solution procedure outlined in this paper will still prove applicable, as long as the reaction rate in the vicinity of the maximum temperature is exponentially large as compared to the reaction rate at lower temperatures [1].

As boundary conditions for the system, it is assumed that far away from the reaction front, no gradients in the mass fluxes of the reacting species or temperature occur:

$$\begin{aligned} x \rightarrow -\infty: \quad & y_k = y_k^u, y_i = y_i^u, T = T^u, \\ & \frac{\partial y_k}{\partial x} = \frac{\partial y_i}{\partial x} = \frac{\partial T}{\partial x} = 0, \\ x \rightarrow \infty: \quad & \frac{\partial y_k}{\partial x} = \frac{\partial y_i}{\partial x} = \frac{\partial T}{\partial x} = 0. \end{aligned} \quad (7)$$

The choice of the boundary conditions (7) reveals the so called ‘‘cold boundary’’ difficulty [4, 18–20]. Activation energy asymptotics elegantly

circumvents the improperly posed problem [20]. The basic idea of the method is that the mixture ahead of any reaction front is reacting exponentially slow and may therefore be ignored for times that are not correspondingly large. The asymptotic analysis, which will be presented in the next section, applies for such times.

DIMENSIONLESS FORMULATION

As customary in applying activation energy asymptotics, a coordinate transformation is performed in which the temperature T is taken as independent variable and the diffusive heat flux q as dependent variable:

$$q = \Lambda_{a,eff} \frac{\partial T}{\partial x} \rightarrow \Lambda_{a,eff} \frac{\partial}{\partial x} = q \frac{\partial}{\partial T} \quad (8)$$

Variables are nondimensionalized as

$$\begin{aligned} T &= T^b - (T^b - T^u)\theta \\ y_k &= y_k^b - (y_k^b - y_k^u)\sigma \\ y_i &= y_i^b - (y_i^b - y_i^u)\zeta \end{aligned} \quad (9)$$

$$q = q_d \psi$$

where the dimensionalization of the diffusive heat q_d is defined by

$$\begin{aligned} q_d^2 &= \Lambda_{a,eff} (-\Delta H_r) k_o e^{-E_a/RT^b} \rho_s^m \left(\frac{\rho_g T^u}{T^b} \right)^n \\ &\cdot (y_k^u - y_k^b)^m (y_i^u - y_i^b)^n (T^b - T^u). \end{aligned} \quad (10)$$

Upon substituting the above relations into Eqs. 1–3 and nondimensionalizing, the nondimensional conservation equations are obtained. Making use of the total energy balance, which is obtained by integrating Eq. 3 over the reaction front under the boundary conditions (7), the following equations for the scaled mass fraction of solid key reactant σ , the scaled mass fraction of gaseous key reactant ζ , and the dimensionless diffusive heat flux ψ are obtained:

$$\psi \frac{\partial}{\partial \theta} \left(\psi \frac{1}{\text{Le}_s} \frac{\partial \sigma}{\partial \theta} \right) + \phi \psi \frac{\partial \sigma}{\partial \theta} - r(\theta, \sigma, \zeta) = 0, \quad (11)$$

$$\psi \frac{\partial}{\partial \theta} \left(\psi \frac{1}{\text{Le}_g} \frac{\partial \zeta}{\partial \theta} \right) + \phi \psi \frac{\partial \zeta}{\partial \theta} - r(\theta, \sigma, \zeta) = 0, \quad (12)$$

$$\psi \frac{\partial \psi}{\partial \theta} + \phi \psi - r(\theta, \sigma, \zeta) = 0, \quad (13)$$

where $r(\theta, \sigma, \zeta)$ is the dimensionless effective reaction rate defined as

$$\begin{aligned} r(\theta, \sigma, \zeta) &= \frac{e^{-(\theta/\gamma)/(1-\mu\theta)}}{1 + \frac{1}{K_g} e^{-(\theta/\gamma)/(1-\mu\theta)}} (\epsilon_k + \sigma)^m \\ &\cdot (\epsilon_i + \zeta)^n \left(\frac{1}{1 - \mu\theta} \right)^n. \end{aligned} \quad (14)$$

A linear temperature dependency of the gas density has been assumed, which is correct in first-order approximation.

The parameters Le_s and Le_g are effective Lewis numbers for solid and gaseous reactant, respectively,

$$\begin{aligned} \text{Le}_s &= \frac{\Lambda_{a,eff}}{\left(\rho_s C_{Ps} + \frac{u_g}{u_f} \rho_g C_{Pg} \right) E_s}, \\ \text{Le}_g &= \frac{\Lambda_{a,eff}}{\left(\rho_g C_{Pg} + \frac{u_f}{u_g} \rho_s C_{Ps} \right) D_{a,eff}}, \end{aligned} \quad (15)$$

while ϕ is dimensionless convective energy flux,

$$\phi = \frac{(u_g \rho_g C_{Pg} + u_f \rho_s C_{Ps})(T^b - T^u)}{q_d} \quad (16)$$

whereby it is noted that ϕ or alternatively the reaction front velocity u_f , is an unknown quantity which follows from solving Eqs. 11–13 subject to appropriate boundary conditions.

The ratio

$$K_g = \frac{k_g}{k_o e^{-E_a/RT^b}} \quad (17)$$

compares external reaction to internal reaction kinetics at temperature T^b ;

$$\mu = \frac{T^b - T^u}{T^b}, \quad \epsilon_i = \frac{y_i^b}{y_i^u - y_i^b}, \quad \epsilon_k = \frac{y_k^b}{y_k^u - y_k^b}, \quad (18)$$

represent relative temperature rise and relative gas and solid conversions, respectively; and

$$\gamma = \frac{RT_b^2}{E_a(T^b - T^u)} \quad (19)$$

is the Zeldovich number. The boundary conditions are

$$\theta = 1(x \rightarrow -\infty): \quad \psi = 0, \quad \sigma = 1, \quad \zeta = 1, \quad (20)$$

$$\theta = 0(x \rightarrow \infty): \quad \psi = 0, \quad \sigma = 0, \quad \zeta = 0. \quad (21)$$

In this analysis we are concerned with the situation where either the gaseous or the solid reactant has converted completely as $\theta = 0$ ($x \rightarrow \infty$). Accordingly, $r = 0$ at $\theta = 0$, implying (cf. Eq. 14) that either ϵ_k or ϵ_i is zero, corresponding to the cases of oversupply of solid and gaseous reactant, respectively.

Large activation energy asymptotics is involved with small values of the Zeldovich number γ :

$$\gamma \ll 1. \quad (22)$$

Distinction can then be made between the preheat zone or outer zone $\theta = 0(1)$ and the reaction zone or inner zone $\theta = 0(\gamma)$. Solutions for these areas using principles of matched asymptotic expansions are presented in the subsequent sections.

SOLUTIONS FOR THE PREHEAT (OUTER) ZONE

In the preheat zone, the reaction term is exponentially small: Eqs. 10–12 can be written as

$$\psi_o^{ou} \frac{\partial}{\partial \theta} \left(\psi_o^{ou} \frac{1}{\text{Le}_s} \frac{\partial \sigma_o^{ou}}{\partial \theta} \right) + \phi \psi_o^{ou} \frac{\partial \sigma_o^{ou}}{\partial \theta} = 0, \quad (23)$$

$$\psi_o^{ou} \frac{\partial}{\partial \theta} \left(\psi_o^{ou} \frac{1}{\text{Le}_g} \frac{\partial \zeta_o^{ou}}{\partial \theta} \right) + \phi \psi_o^{ou} \frac{\partial \zeta_o^{ou}}{\partial \theta} = 0, \quad (24)$$

$$\psi_o^{ou} \frac{\partial \psi_o^{ou}}{\partial \theta} + \phi \psi_o^{ou} = 0, \quad (25)$$

where ψ_o^{ou} , σ_o^{ou} , and ζ_o^{ou} correspond to the first terms of an expansion involving γ of the solutions of ψ , σ , and ζ in the preheat or outer zone, respectively.

The solution of Eq. 25 satisfying boundary condition (20) is easily obtained as

$$\psi_o^{ou} = \phi(1 - \theta). \quad (26)$$

Substituting this result into Eqs. 23 and 24 and solving subject to boundary conditions (20) one obtains

$$\sigma_o^{ou} = 1 + \frac{B_0}{\text{Le}_s} (1 - \theta)^{\text{Le}_s}, \quad (27)$$

$$\zeta_o^{ou} = 1 + \frac{C_0}{\text{Le}_g} (1 - \theta)^{\text{Le}_g}, \quad (28)$$

where the constants B_0 and C_0 follow from matching the solutions to those appropriate for the inner or reaction zone.

SOLUTIONS FOR THE REACTION (INNER) ZONE

To derive a solution appropriate for the inner zone, a new independent variable, the inner coordinate η is introduced:

$$\eta = \frac{\theta}{\gamma} \rightarrow \frac{\partial}{\partial \theta} = \frac{1}{\gamma} \frac{\partial}{\partial \eta} \quad (29)$$

Furthermore, a balance of terms which is physically meaningful for the reaction zone becomes apparent if we scale σ , ζ , and ψ as

$$\sigma = \sigma^{in} \quad (30)$$

$$\zeta = \gamma \text{Le}_g \zeta^{in}$$

$$\psi = \gamma^{(n+1)/2} (\text{Le}_g)^{n/2} \psi^{in}$$

and the dimensionless convective energy flux ϕ as

$$\phi = \gamma^{n+1/2} (\text{Le}_g)^{n/2} \phi_o \quad (31)$$

Equations 10–12 then become

$$\psi^{in} \frac{\partial}{\partial \eta} \left(\frac{\psi^{in}}{\gamma \text{Le}_s} \frac{\partial \sigma^{in}}{\partial \eta} \right) + \phi_o \psi^{in} \frac{\partial \sigma^{in}}{\partial \eta} - r_o(\theta^{in}, \sigma^{in}, \zeta^{in}) = 0 \quad (32)$$

$$\psi^{in} \frac{\partial}{\partial \eta} \left(\psi^{in} \frac{\partial \zeta^{in}}{\partial \eta} \right) + \gamma \text{Le}_g \phi_o \psi^{in} \frac{\partial \zeta^{in}}{\partial \eta} - r_o(\theta^{in}, \sigma^{in}, \zeta^{in}) = 0 \quad (33)$$

$$\psi^{in} \frac{\partial \psi^{in}}{\partial \eta} + \gamma \phi_o \psi^{in} - r_o(\theta^{in}, \sigma^{in}, \zeta^{in}) = 0 \quad (34)$$

where

$$r_o(\theta^{in}, \sigma^{in}, \zeta^{in}) = (\gamma L_e g)^{-n} r(\theta^{in}, \sigma^{in}, \gamma L_g \zeta^{in}) \quad (35)$$

is the scaled dimensionless reaction rate.

Clearly, the scaling of variables has been chosen such that for $\gamma \rightarrow 0$, a balance occurs between heat of conduction and heat of reaction in heat transport equation (34). The situation in the equations pertaining to transport of solid and gaseous reactants, cf. Eqs. 32 and 33, depends on the magnitude of the Lewis numbers Le_s and Le_g with respect to the magnitude of γ . Guided by values encountered in practice, the effective diffusivity of the solid-phase reactant E_s is taken to be very low such that

$$\gamma Le_s \gg 1. \quad (36)$$

The effective Lewis number of the gaseous phase is such that

$$\gamma Le_g \ll 1 \quad (37)$$

Denoting the first term of an expansion involving γ , γLe_g , and $(\gamma Le_s)^{-1}$ in the solutions for ψ^{in} , ζ^{in} , and σ^{in} by the subscript o , we have from Eqs. 32–35:

$$\phi_o \psi_o^{in} \frac{\partial \sigma_o^{in}}{\partial \eta} - r_o(\theta_o^{in}, \sigma_o^{in}, \zeta_o^{in}) = 0, \quad (38)$$

$$\psi_o^{in} \frac{\partial}{\partial \eta} \left(\psi_o^{in} \frac{\partial}{\partial \eta} \zeta_o^{in} \right) - r_o(\theta_o^{in}, \sigma_o^{in}, \zeta_o^{in}) = 0, \quad (39)$$

$$\psi_o^{in} \frac{\partial \psi_o^{in}}{\partial \eta} - r_o(\theta_o^{in}, \sigma_o^{in}, \zeta_o^{in}) = 0, \quad (40)$$

where

$$\frac{\psi_o^{in} (\epsilon_k \phi_o + \psi_o^{in})^{1-m}}{1-m} - \frac{(\epsilon_k \phi_o + \psi_o^{in})^{2-m}}{(1-m)(2-m)} + \frac{(\epsilon_k \phi_o)^{2-m}}{(1-m)(2-m)} = \frac{K_g}{\phi_o^m} \int_o^\eta \frac{(\alpha + \eta)^n e^{-\eta}}{e^{-\eta} + K_g} d\eta \quad (m \neq 1, 2), \quad (45)$$

$$\psi_o^{in} - \epsilon_k \phi_o \ln(\epsilon_k \phi_o + \psi_o^{in}) + \epsilon_k \phi_o \ln(\epsilon_k \phi_o) = \frac{K_g}{\phi_o^m} \int_o^\eta \frac{(\alpha + \eta)^n e^{-\eta}}{e^{-\eta} + K_g} d\eta \quad (m = 1),$$

$$\ln(\epsilon_k \phi_o + \psi_o^{in}) - \frac{\psi_o^{in}}{\epsilon_k \phi_o + \psi_o^{in}} - \ln(\epsilon_k \phi_o) = \frac{K_g}{\phi_o^m} \int_o^\eta \frac{(\alpha + \eta)^n e^{-\eta}}{e^{-\eta} + K_g} d\eta \quad (m = 2),$$

Equation 45 is an implicit equation, from which the 1-term inner solution of the dimensionless diffusive heat flux ψ as function of the

$$r_o(\theta_o^{in}, \sigma_o^{in}, \zeta_o^{in}) = \frac{e^{-\eta}}{1 + \frac{1}{K_g} e^{-\eta}} (\epsilon_k + \sigma_o^{in})^m (\alpha_i + \zeta_o^{in})^n. \quad (41)$$

and

$$\alpha_i = \epsilon_{ij} / (\gamma Le_g). \quad (42)$$

As any gaseous reactant surplus is considered to be small, the parameter α_i is assumed to be 0(1) at most.

Eliminating the reaction term from Eqs. 39 and 40 and from Eqs. 38 and 40, respectively, integrating the resulting equations, and applying boundary condition (21) yields

$$\zeta_o^{in} = \eta \quad (43)$$

$$\sigma_o^{in} = \frac{\psi_o^{in}}{\phi_o}$$

The equation determining ψ_o^{in} then follows from Eqs. 40–43 as

$$\psi_o^{in} \frac{\partial \psi_o^{in}}{\partial \eta} - K_g \left(\epsilon_k + \frac{\psi_o^{in}}{\phi_o} \right)^m (\alpha_i + \eta)^n \frac{e^{-\eta}}{e^{-\eta} + K_g} = 0 \quad (44)$$

The solution to this equation is found by integration, where distinction has to be made between the cases $m \neq 1, 2$; $m = 1$, and $m = 2$, respectively:

boundary coordinate η can be solved by integrating. Unfortunately, an analytic solution of the integral in Eq. 45 can not be derived for

general values of the parameter K_g and general reaction orders n . For first-order reactions ($n = 1$), it is known that the resulting integral can not be written as a finite combination of elementary functions [9]. In all cases, the integral is convergent and can be solved numerically. In some specific cases, it is possible to solve the integral in closed form. These cases are: general Arrhenius kinetics without transport limitations ($K_g \rightarrow \infty$), general values of K_g in combination with zero-th order reactions ($n = 0$), and the case $K_g = 1$ for general reaction orders [1].

For the important special case $\epsilon_k = 0$, an explicit solution in terms of an integral can be obtained ($m < 2$):

$$\psi_o^{in} = \left(\frac{2-m}{\phi_o^m} K_g \int_o^\eta \frac{(\alpha_i + \eta)^n e^{-\eta}}{e^{-\eta} + K_g} d\eta \right)^{1/2-m} \quad (46)$$

This solution depicts understoichiometric reaction with respect to the reactant y_k , where $y_k^b = 0$.

MATCHING INNER AND OUTER SOLUTIONS

The dimensionless convective energy flux ϕ follows from matching the inner solution for the dimensionless diffusive heat flux ψ to the outer solution [2]. This approach can be performed easily in case $\epsilon_k = 0$, because an explicit solution is known. If $\epsilon_k \neq 0$, the 1-term outer expansion of the 1-term inner solution can be found by taking the limit for $\eta \rightarrow \infty$ in Eq. 45 and

solving for ψ . The formal matching procedure corresponds to equating the limit-value for $\theta \rightarrow 0$ of the 1-term outer solution to the limit-value for $\eta \rightarrow \infty$ of the 1-term inner solution.

For reasons of illustration, the case $\epsilon_k = 0$ will be worked out, since an explicit solution for ψ_o^{in} can be obtained for this situation. The 1-term outer solutions for the scaled mass fractions σ and ζ and the dimensionless diffusive heat flux ψ are given by Eqs. 26, 27, and 28, respectively. The 1-term inner solutions are given by Eqs. 30, 31, 43, and 46. The matching procedure is performed for the dimensionless diffusive heat flux ψ first, yielding the convective energy flux ϕ . The result is substituted in the inner solution for the scaled mass fraction of the key reactant in the solid phase σ , followed by matching σ and ζ , yielding the integration constants B_0 and C_0 .

The result of the procedure is given by:

$$\begin{aligned} B_0 &= 0, \\ C_0 &= -Le_g, \\ \phi &= \gamma^{n+1/2} Le_g^{n/2} \phi_o, \end{aligned} \quad (47)$$

$$\phi_o = \sqrt{(1-m)K_g \int_o^\infty \frac{(\alpha_i + \eta)^n e^{-\eta}}{e^{-\eta} + K_g} d\eta}$$

Within the validity range of the solution, the following composite solution for the dimensionless mass fractions σ and ζ and diffusive flux ψ can be constituted by applying the matching principle [2]:

$$\begin{aligned} \sigma(\theta) &= \left(\frac{\int_o^{\theta/\gamma} \frac{(\alpha_i + \eta)^n e^{-\eta}}{e^{-\eta} + K_g} d\eta}{\int_o^\infty \frac{(\alpha_i + \eta)^n e^{-\eta}}{e^{-\eta} + K_g} d\eta} \right)^{1/2-m}, \\ \zeta(\theta) &= 1 - (1-\theta)^{Le_g}, \\ \psi &= \gamma^{n+1/2} Le_g^{n/2} \psi_o(\theta), \\ \psi_o(\theta) &= \left(\frac{(2-m)K_g}{\left((2-m)K_g \int_o^\infty \frac{(\alpha_i + \eta)^n e^{-\eta}}{e^{-\eta} + K_g} d\eta \right)^{m/2}} \int_o^{\theta/\gamma} \frac{(\alpha_i + \eta)^n e^{-\eta}}{e^{-\eta} + K_g} d\eta \right)^{1/2-m} \\ &\quad - \theta \left((2-m)K_g \int_o^\infty \frac{(\alpha_i + \eta)^n e^{-\eta}}{e^{-\eta} + K_h} d\eta \right)^{1/2}. \end{aligned} \quad (48)$$

Equations 47 and 48 are in essence a very complex set of solutions, in which many situations are described. In order to illustrate the importance of these, two simplified cases will be considered here in more depth. Other examples are described elsewhere [1].

RESULTS FOR $m = 0$ AND $K_G \rightarrow \infty$

In the limit $m \rightarrow 0$, there is no dependency of the reaction rate on the concentration of the solid key reactant y_k . In that situation, the solid mass balance (11) can be disregarded. As has been remarked, the integral appearing in Eqs.

$$\psi = \gamma^{n+1/2} \text{Le}_g^{n/2} \psi_o(\theta), \quad (50)$$

$$\psi_o(\theta) = \sqrt{2e^{\alpha_i}(\Gamma((n+1), (\alpha_i + \theta/\gamma)) - \Gamma((n+1), \alpha_i)) - \theta \sqrt{2e^{\alpha_i}(\Gamma(n+1) - \Gamma((n+1), \alpha_i))}}$$

For general positive reaction orders n , the incomplete gamma function Γ appears in the solution [21]. In the special case of homogeneous combustion, e.g., combustion of gases in premixed flames ($\epsilon_g = 1$), and for $\text{Le}_g = 1$ and $\alpha_i = 0$ (i.e., the gaseous key reactant is fully consumed) and the reaction order n being a natural number, the above solution reduces to that found by Zeldovich, Frank-Kamenetsky, and Semenov [4, 18, 19, 22] and by Merzhanov [5]. The case $\alpha_i = 0$ can be easily obtained in premixed flames, by taking the reactant that is fully consumed as the gaseous key reactant y_i . Similar solutions for general Lewis numbers of order unity and $\alpha_i = 0$ have been obtained, making use of activation energy asymptotics, by Bush and Fendell [4] and Zeldovich et al. [23]. As far as known to the authors, the situation $\alpha_i > 0$ (in which the gaseous key reactant is in excess) has not been solved before. In the following pages, results for reaction front propagation with dominant diffusive transport of the reacting species in the reaction zone for true Arrhenius kinetics will be presented graphically.

The matching procedure is illustrated by Fig. 2, where the inner, outer, and composite solutions for the dimensionless diffusive heat flux ψ_o are given as function of dimensionless temperature θ , for a first-order reaction, $\alpha_i = 0$ and $\gamma = 0.1$ and $\gamma = 0.01$ respectively. For smaller values of the parameter γ , the reaction zone

47 and 48 can be solved analytically if $K_g \rightarrow \infty$, which describes a true Arrhenius type of reaction (no transport limitation of the chemical reaction). The following expressions are obtained for the dimensionless convective energy flux ϕ , the scaled mass fraction ζ , and the dimensionless diffusive heat flux ψ

$$\phi = \gamma^{n+1/2} \text{Le}_g^{n/2} \phi_o, \quad (49)$$

$$\phi_o = \sqrt{2e^{\alpha_i}(\Gamma(n+1) - \Gamma((n+1), \alpha_i))}$$

$$\zeta(\theta) = 1 - (1 - \theta)^{\text{Le}_g},$$

(located at $\theta \rightarrow 0$) is shifted to smaller values of θ . In other words, the boundary layer where the inner solution is valid becomes smaller. As is expected from the exponential temperature dependence of the reaction rate, for a higher activation energy, a larger part of the conversion occurs in the vicinity of the maximum temperature T^b (where $\theta = 0$).

The influence of parameter α_i on the dimensionless convective energy flux ϕ_o (and consequently on the propagation speed of the reaction front) is illustrated in Fig. 3, for various values of the reaction order n . It can be seen that ϕ_o increases with increasing α_i . The param-

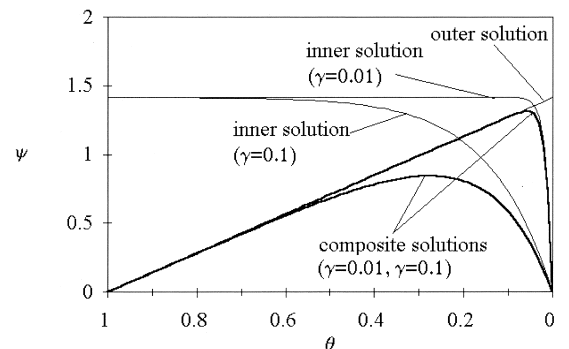


Fig. 2. Inner, outer, and composite solutions for the dimensionless diffusive energy flux ψ_o , for $\gamma = 0.1$ and $\gamma = 0.01$ ($n = 1$, $\alpha_i = 0$).

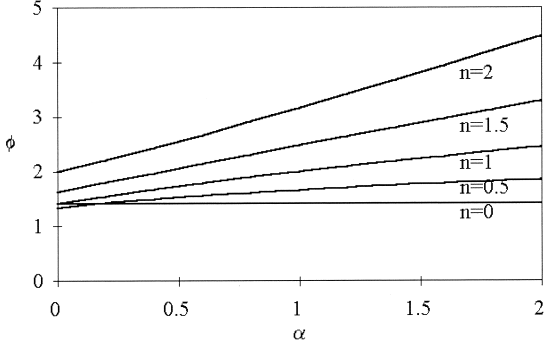


Fig. 3. Dimensionless convective energy flux ϕ_o as function of parameter α_i , for various reaction orders.

eter α_i is larger than zero for overstoichiometric supply of the gaseous reactant (the gaseous reactant is in excess). This results in a higher reaction rate at the maximum combustion temperature, caused by the larger concentration of the gaseous reactant in the reaction zone. The maximum temperature is limited by the supply of the solid reactant. The composite solution for the dimensionless mass fraction ζ as function of the dimensionless temperature θ is given in Fig. 4 for various values of the effective Lewis number. The parameters n , α_i , and γ have no influence on the composite solution as a function of θ . If $Le_g = 1$, diffusion of species occurs at equal speed as heat conduction (equi-diffusion [20]). For $Le_g > 1$, the conduction of heat is faster than diffusion of species. The concentration of the reacting species in the reaction zone is found to increase with increasing Lewis numbers, which is reflected in an increase in the reaction front velocity u_f .

Differentiating the expression for T in Eq. 9

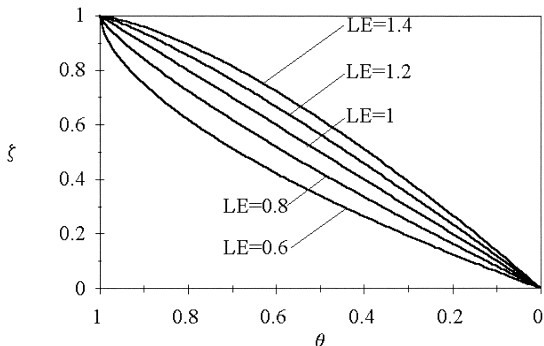


Fig. 4. Composite solution for the dimensionless mass fraction ζ for various Lewis numbers.

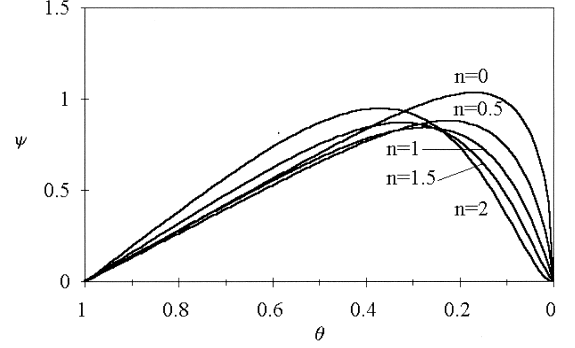


Fig. 5. Composite solution for the dimensionless heat flux ψ_o for various reaction orders ($\gamma = 0.1$, $\alpha_i = 0$).

and substituting into Eq. 8, making use of the definition of y in Eq. 9 and the relation between y and y_o in Eq. 50, the solution for the dimensionless mass fraction ζ is only a function of the effective Lewis number. The solution for the dimensionless diffusive heat flux ψ_o , however, depends on the reaction order n , as well as on the parameters α_i and γ . The influence of the magnitude of γ on the solution for a first-order reaction was already shown in Fig. 2. The influence of the reaction order is illustrated by Fig. 5, for the case $\gamma = 0.1$ and $\alpha_i = 0$. As the extent of the reaction zone increases with increasing reaction order, the maximum of ψ_o shifts to the direction of $\theta = 1$ with increasing reaction order.

The following relation between ψ_o and θ can be derived:

$$\psi_o = -\frac{\partial \theta}{\partial X}, \quad X = \frac{q_d \gamma^{n+1/2} Le_g^{n/2} x}{\Lambda_{a,eff}(T^b - T^u)}. \quad (51)$$

Upon equating with the first-order solution for ψ given by Eq. 50 and integrating once, the distribution of dimensionless temperature as a function of distance X is obtained: $\theta = \theta(X)$. The integration has to be performed numerically. Substituting the result in Eq. 50 finally gives the dimensionless diffusive heat flux ψ and dimensionless mass fraction ζ as function of coordinate X .

Figure 6 shows an example of this procedure. Here, the dimensionless temperature θ and mass fraction ζ have been presented as a function of the scaled distance X for the case of a first-order reaction, with $\gamma = 0.1$ and $\alpha_i = 0$. As the scaled distance X is dependent on the Lewis

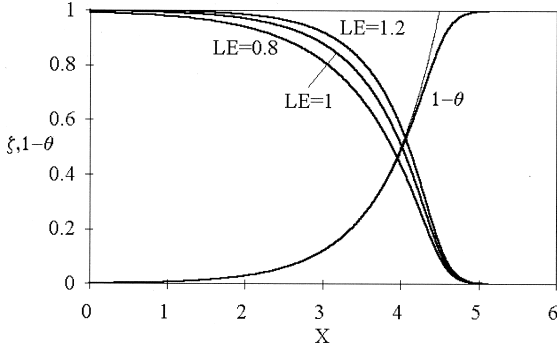


Fig. 6. Dimensionless temperature θ and mass fraction ζ for various Lewis number ($n = 1$, $\gamma = 0.1$, $\alpha_i = 0$).

number, Fig. 6 must be interpreted carefully. The relative location of the mass fraction when compared to the dimensionless temperature, however, can be clearly observed. The thin curve shows the extrapolation of the outer solution for the dimensionless diffusive heat flux ψ . Comparing the composite solution to the outer solution gives an estimate of the extent of the reaction zone. The results for $\gamma = 0.01$ are similar to Fig. 6 and will not be presented here. The main difference is a thinner reaction layer as function of the scaled distance X , as might be expected from the stronger dependence of the reaction rate on temperature.

RESULTS FOR $n = 0$ AND $K_G \rightarrow \infty$

In the limit $n \rightarrow 0$, there is no dependency of the reaction rate on the concentration of the gaseous key reactant y_i . Similarly to the previous paragraph, solutions for the remaining set of equations can be obtained. In the limit $K_g \rightarrow \infty$, the analysis simplifies considerably and will be treated here to enable comparison with the previous paragraph. After some mathematical derivations, Eqs. 47 and 48 reduce to:

$$\phi = \gamma^{1/2} \phi_o, \quad \phi_o = \sqrt{2 - m}, \quad (52)$$

$$\sigma(\theta) = (1 - e^{-\theta/\gamma})^{1/(2-m)}, \quad (53)$$

$$\begin{aligned} \psi &= \gamma^{1/2} \psi_o, \quad \psi_o(\theta) \psi \\ &= \gamma^{1/2} \psi_o, \quad \psi_o(\theta) \\ &= \sqrt{2 - m} (1 - e^{-\theta/\gamma})^{1/(2-m)} - \sqrt{2 - m} \theta. \end{aligned}$$

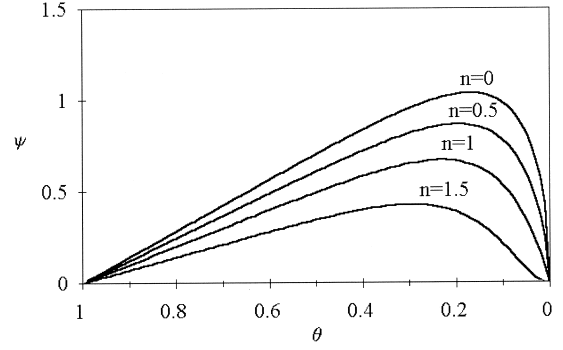


Fig. 7. Composite solution for the dimensionless heat flux ψ_o for various reaction orders ($\gamma = 0.1$).

Figures 7 and 8 show the composite solutions for the dimensionless diffusive heat flux ψ_o and the dimensionless mass fraction σ respectively. The parameter along the various curves is the reaction order m and the parameter γ is taken as $\gamma = 0.1$.

As before, the thickness of the reaction zone decreases with decreasing γ . Furthermore, for higher order reactions, the reaction zone is shifted toward higher concentrations of the reactant. This causes the maximum for ψ_o to shift to the left.

The scaled mass fraction σ is equal to 1 in the preheat zone, as can be observed from Fig. 8. The first decrease occurs at the point where reaction is started. The extent of the reaction zone decreases with the parameter γ and the reaction order n , corresponding to the findings for the dimensionless diffusive heat flux ψ_o .

As before, the composite solution for ψ can be integrated, yielding an expression for the dimensionless temperature θ , as function of a

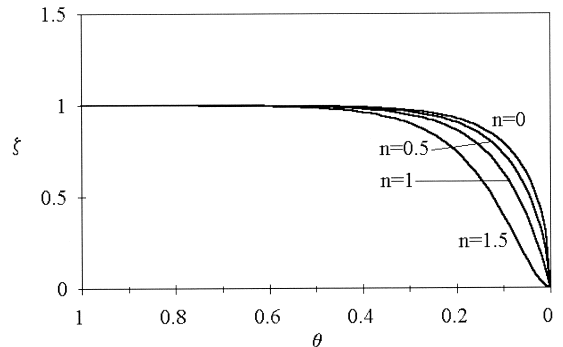


Fig. 8. Composite solution for the dimensionless mass fraction σ , for various reaction orders ($\gamma = 0.1$).

scaled distance X . Integration has to be done numerically. Results are given elsewhere [1].

EFFECTS OF LEWIS NUMBER

The results discussed in the previous two sections apply to reaction front propagation with a single key reactant, i.e., the gas phase as key reactant obtained by setting $m = 0$ and the solid phase as key reactant obtained by setting $n = 0$ in general solutions (47) and (48). In obtaining these solutions the Lewis number for the gas phase was taken such that $\gamma Le_g \ll 1$ so that convective transport of gaseous reactant in the reaction zone could be disregarded: cf. Eq. 33. Conversely, for the solid phase the Lewis number was taken such that $\gamma Le_s \gg 1$, with the result that diffusive transport of solid reactant in the reaction zone could be disregarded: cf. Eq. 32. As the mathematical expressions for the dependencies on solid and gaseous reactant are similar, the results of the previous two sections can be treated as the solutions for reaction front propagation involving a single key reactant for the limits of $\gamma Le \ll 1$ and $\gamma Le \gg 1$, respectively. By comparing these solutions the effect of the Lewis number on reaction front propagation can be established.

From solution (50) it follows that in case of dominant diffusive transport of the reacting species in the reaction zone, i.e., when $\gamma Le \ll 1$, the concentration of the reacting species at $\theta \sim \gamma$, i.e., in the reaction zone, γLe . Conversely, in case of dominant convective transport in the reaction zone, i.e., when $\gamma Le \gg 1$, it follows from solution (53) that the concentration of reacting species is of unit order of magnitude. From solutions (49) and (52) it follows that the dimensionless convective energy flux ϕ which determines the reaction front velocity (cf. Eq. 16), $\sim \gamma^{n+1/2} Le^{n/2}$ when $\gamma Le \ll 1$ and $\sim \gamma^{1/2}$ when $\gamma Le \gg 1$.

When $\gamma Le = O(1)$, both convection and diffusion of reacting species are important in the reaction zone. As for this situation no simplification of the mass balance equation is possible, finding solutions from the governing equations becomes a cumbersome task. The solutions for this situation will have characteristics of the two limiting cases. Therefore, it is

proposed to construct an "engineering solution" that provides a smooth change-over from the one limiting solution to the other solution, as function of the Lewis number. An important parameter for overall engineering models is the reaction front propagation speed. As indicated below, this overall parameter can be described by such an approach. Combining the different temperature and concentration distributions of the two limit situations is not realizable.

When the solutions for the dimensionless convective energy flux are denoted by the superscripts d and c for the diffusion ($\gamma Le \ll 1$) and convection dominated ($\gamma Le \gg 1$) limits, we have

$$\frac{\phi^d}{\phi^c} = \frac{\phi_o^d}{\phi_o^c} (\gamma Le)^{n/2} \quad (54)$$

where ϕ_o^d and ϕ_o^c are $O(1)$ quantities. In case $\gamma Le \ll 1$, the solution is given by ϕ^d . When $\gamma Le \gg 1$, the solution is captured by ϕ^c . On basis of this result, the following approximate solution can be constructed for general values of Le :

$$\phi^{\text{appr.}} = \left(\frac{1}{\frac{1}{(\phi^d)^\delta} + \frac{1}{(\phi^c)^\delta}} \right)^{1/\delta}, \quad (55)$$

where δ is an arbitrary but positive constant. In the limiting values, the solution goes to ϕ^d when $\gamma Le \ll 1$ and ϕ^c when $\gamma Le \gg 1$. The value of δ can be assessed by adjusting to numerical or experimental results.

Fortunately, most practical situations are well described by one of the limiting cases. As an example, the premixed combustion of gases in laminar flames can be mentioned, where $Le = O(1)$. Furthermore, combustion synthesis processes in a solid phase are generally characterized by $Le \rightarrow \infty$. For heterogeneous combustion in packed beds, Le_g appears to be of $O(1)$, whereas the solid-phase dispersion is characterized by very large Lewis numbers. For this situation, the general solutions as described by Eqs. 47 and 48 can be applied.

Finally, it is important to make a final remark on the choice of the small parameter γ :

$$\gamma = \frac{R(T^b)^2}{E_{act}(T^b - T^u)} = \frac{T^b}{R/E_{act}} \frac{T^b}{T^b - T^u}. \quad (56)$$

According to Eq. 56, the requirement $\gamma \ll 1$ has two implications. First, the activation temperature (R/E_{act}) has to be large as compared to the maximum reaction temperature T^b . Furthermore, the temperature ($T^b - T^u$) must be of the same order as the maximum reaction temperature T^b , i.e., the initial temperature is required to be much smaller than the final reaction temperature. It follows that the propagation of a reaction front is related to a strong temperature dependence of the reaction rate and also to a large reaction heat. The requirement $\gamma \ll 1$ is more severe than the requirement $\epsilon = RT^b/E_a \ll 1$, used by other authors [20, 23–25]. This parameter can be much smaller than unity even when the difference ($T^b - T^u$), i.e., the adiabatic temperature rise, is small. If the latter is not the case, however, the reaction zone may well be non-narrow and reaction will proceed over the entire domain. This may occur in situations where the reactants have to be heated to a temperature close to the maximum reaction temperature, before entering the reactor, in order to achieve the necessary conversion. In this case an asymptotic treatment based on conversion in a small boundary layer is no longer applicable.

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